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Enhanced tunnel magnetoresistance in granular nanobridges

K. Yakushiji,^{a)} S. Mitani, K. Takanashi, S. Takahashi, and S. Maekawa

Institute for Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, 980-8577, Japan

H. Imamura

Graduate School of Information Sciences, Tohoku University, Aramaki, Aoba-ku, Sendai 980-8579, Japan

H. Fujimori

The Research Institute for Electric and Magnetic Materials, 2-1-1 Yagiyama-minami, Taihaku-ku, Sendai, 982-0807, Japan

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We have fabricated granular nanobridge structures consisting of electrodes separated by a nanometer-sized gap in which a thin insulating CoAlO granular film is filled, and measured the current–bias voltage characteristics in a magnetic field to investigate the spin-dependent transport. The Coulomb blockade with a clear threshold voltage (V_{th}) is observed at 4.2 K. Tunnel magnetoresistance (TMR) is enhanced by fabricating nanobridges. TMR shows a maximum exceeding about 30% at the voltage slightly above V_{th} . This enhancement is explained by the orthodox theory of single electron tunneling in ferromagnetic multiple tunnel junctions. © 2001 American Institute of Physics. [DOI: 10.1063/1.1341231]

Interplay of spin-dependent tunneling and electrical charging effect in small metallic particles gives rise to remarkable magnetotransport phenomena. The charging effect of particles leads to single electron tunneling (SET)¹ represented by the Coulomb blockade of electric current below a threshold voltage and Coulomb staircase at higher voltages. The basic structure for the study of SET phenomena consists of electrodes and small islands, which are separated from the electrodes by tunnel barriers (multiple junction). Theoretically, the spin-dependent SET phenomena have been studied by using a multiple junction model with spin-dependent tunnel resistances. The enhancement and oscillation of tunnel magnetoresistance (TMR) in ferromagnetic multiple junctions have been predicted by several authors.^{2–5} Experimentally spin-dependent SET was investigated in microfabricated samples consisting of Ni/NiO/Co/NiO/Ni double tunnel junctions at very low temperatures (~ 20 mK).⁶ However, the microfabrication technique, such as electron-beam lithography, is usually limited to the formation of islands with submicron sizes, and SET phenomena could be expected only at low temperatures. Therefore, the use of nanometer-sized particles naturally formed by self-assembling process has attracted attention^{7–9} to observe SET phenomena at elevated temperatures.

Insulating granular films consisting of nanometer-sized magnetic metallic particles embedded in an insulating matrix are useful for the study of spin-dependent SET phenomena. Insulating granular films exhibit large TMR,¹⁰ and the size of particles is so small that the charging energy reaches up to several hundreds meV. However, in a granular film with macroscopic size containing a vast number of particles, SET phenomena are averaged out due to the large distribution of particle sizes¹¹ and that of interparticle distances. The current paths should be restricted to observe the SET phenomena. A

simple method to restrict the tunneling paths is to use the scanning tunneling microscopy (STM) technique. The current path on the surface is limited to only one particle just below the STM tip. We observed a clear Coulomb staircase in the current–voltage measurements for CoAlO granular films even at room temperature.¹² An advantageous method for a variety of measurements and applications is to fabricate a device structure consisting of a small part of a granular film with microscopic leads.

In this study, we have fabricated point-shaped electrodes separated by a very narrow lateral gap in which an insulating granular film is filled, which we call “granular nanobridges.” We have measured the current (I)–bias voltage (V_b) characteristics in CoAlO granular nanobridges, and found enhanced TMR due to the Coulomb blockade. We apply the orthodox theory of SET and explain that the enhanced TMR is brought about by the modification of the detailed balance of particle charges by the external magnetic field.^{3,4,12}

A schematic view of a typical sample is shown in Fig. 1. An insulating granular nanobridge was fabricated on a glass substrate as follows: a 15 nm thick NbZrSi amorphous layer was deposited by rf sputtering, and was formed into source and drain electrodes by focused ion beam (FIB) etching using 30 kV gallium ions (Seiko Instruments Inc., SMI 9200). The electrodes separated by a gap with a length (l), i.e., gap separation, of 30 nm and a width (w) of 60 nm are shown in

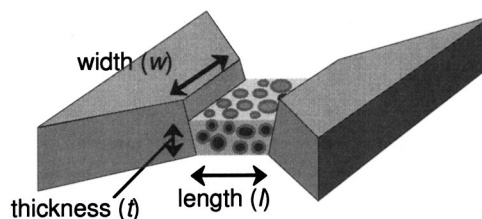


FIG. 1. Schematic view of an insulating granular nanobridge.

^{a)}Electronic mail: kyaku@imr.edu

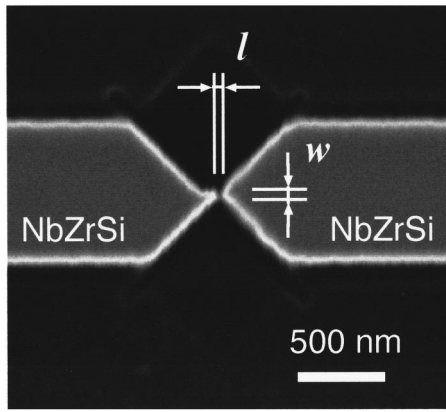


FIG. 2. Scanning ion microscopy image of the NbZrSi electrodes separated by a nanometer-sized lateral gap. The length (l) is 30 nm and the width (w) is 60 nm.

Fig. 2. Deep trenches (60 nm wide and 200 nm deep) were formed beside the gap by FIB etching to avoid the formation of unnecessary current paths outside the gap. A 7.5 nm thick CoAlO granular film was deposited on the patterned surface by reactive rf sputtering, and the gap was filled by the CoAlO film. The aspect ratio of trenches is so high that the trenches are not filled with the CoAlO film. The composition of CoAlO was determined to be $\text{Co}_{36}\text{Al}_{22}\text{O}_{42}$ by Rutherford backscattering analysis. The average size of Co particles was estimated to be about 25 Å from the analysis of the superparamagnetic behavior and the transmission electron microscopy observation.¹¹ The characteristic sizes of granular nano-bridges, i.e., w , l , and thickness (t), were varied in the range of 60–700 nm, 30–70 nm, and 5–30 nm, respectively. I – V_b characteristics were measured at 4.2 K using an electrometer (Keithley 6514) with a two-terminal arrangement. TMR ($=\Delta R/R_{H=0}$) was evaluated from the difference between the I – V_b curves at the applied field $H=0$ and 10 kOe.

Figure 3 (a) shows the I – V_b characteristics at $H=0$ (solid lines) and $H=10$ kOe (dashed lines) for the sample with $w=60$ nm, $l=30$ nm and $t=7.5$ nm. Here, the threshold voltage ($V_{th} \sim 1.5$ V) is defined as that below which the current is zero within the accuracy of 100 fA. In the range of $|V_b| < V_{th}$, the Coulomb blockade occurs. The current increases rapidly when $|V_b|$ exceeds V_{th} . In contrast, V_{th} has not been observed for CoAlO films with macroscopic sizes containing a vast number of Co particles with a broad distribution of sizes,¹¹ and electron tunneling occurs mainly between large particles with small charging energy. On the other hand, in the nanobridge, the tunneling paths are limited and the Coulomb blockade effect is remarkable.

TMR depends strongly on V_b as shown in Fig. 3(b). For $|V_b| < 4.0$ V, TMR increases with decreasing $|V_b|$ and reaches the maximum value larger than 30% at the voltage slightly above V_{th} (~ 1.5 V). For $|V_b| < V_{th}$ (hatching area), there is little quantitative reliability on the measurements because the current in the blockade region is very low (< 100 fA). For $|V_b| > 4.0$ V, on the other hand, TMR shows no large change and converges toward about 8%. Throughout the measured range, the V_b dependence of TMR shows oscillatory behavior, which is possibly related to SET rather than the discrete electronic levels.¹³

Similar results have been obtained in other samples. For

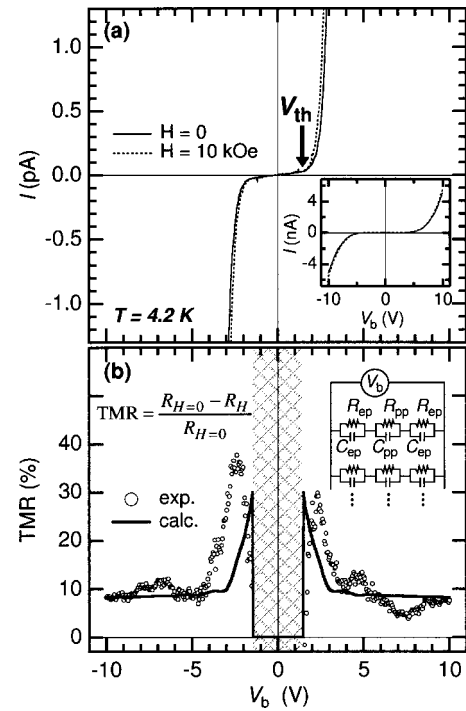


FIG. 3. (a) Current–bias voltage (I – V_b) characteristics and (b) V_b dependence of TMR measured at 4.2 K for the sample with $w=60$ nm, $l=30$ nm, and $t=7.5$ nm. In (a), the solid and dashed curves represent I – V_b curves in a magnified current range at $H=0$ and 10 kOe, respectively. I – V_b curves throughout the measured current range are shown in the inset. V_{th} denotes the Coulomb threshold voltage (~ 1.5 V). In Fig. 3 (b), the result obtained by the numerical calculation is shown in a solid curve. The hatching area represents the Coulomb blockade region ($|V_b| < V_{th}$). The conducting path in the contact is modeled by a parallel circuit of 20 triple-tunnel junctions as shown in the inset of panel (b).

the sample with $w=700$ nm, $l=40$ nm, and $t=15$ nm, V_{th} is observed to be 0.4 V which is lower than that in the sample shown in Fig. 3, suggesting that V_{th} increases with decreasing the sample size. The voltage where the TMR shows a maximum V_p , is slightly larger than V_{th} . Figure 4 shows V_p vs V_{th} in the samples with various sizes. V_p is close to and slightly larger than V_{th} , indicating that the enhanced TMR is caused by the Coulomb blockade.

We apply the orthodox theory of SET¹ and explain the experimental results. The conducting path in the nanobridge is modeled by a parallel circuit of triple-tunnel junctions as shown in the inset of Fig. 3(b). This is the simplest model to

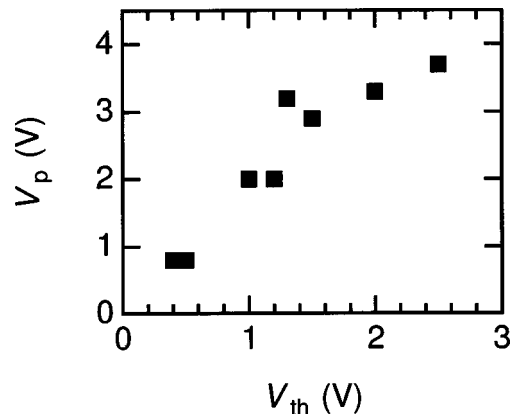


FIG. 4. V_p vs V_{th} for the samples with various sizes. V_p is the voltage where the TMR shows the maximum.

explain our experimental results because we need at least two granules in each series junction to study the spin-dependent transport in nanobridges with nonmagnetic electrodes. We neglect the higher-order tunneling process called cotunneling^{2,14} because the tunnel resistances between particles and between the electrode and a particle are estimated to be about 10^5 times larger than the quantum resistance, $R_K \equiv h/e^2 \approx 26 \text{ k}\Omega$. In order to obtain the stable tunneling current, we construct the detailed balance equation for the probability of states $p(\{n_{ij}\}_\alpha)$, which is given in the matrix form by $\dot{\mathbf{p}} = \mathbf{M}\mathbf{p} = 0$, where $\mathbf{p} = (\dots, p(\{n_{ij}\}_\alpha), \dots)^T$ and \mathbf{M} is the transition matrix in the configuration space constructed by $\{n_{ij}\}_\alpha$ with the index α labeling the different charge states. The tunneling current through the k th junction is given by $I_k = e \sum_\alpha p(\{n_{ij}\}_\alpha) [\Gamma_k^+(\{n_{ij}\}_\alpha) - \Gamma_k^-(\{n_{ij}\}_\alpha)]$, where $\Gamma_k^{+(-)}(\{n_{ij}\}_\alpha) \propto 1/R_k$ is the forward (backward) tunneling rate through the k th junction with the initial charge state $\{n_{ij}\}_\alpha$. The current conservation requires that the tunneling current I_k is the same for all junctions in each series junction. Let us evaluate the tunneling current at the junction between the nonmagnetic electrode and a particle, where the tunneling rate $\Gamma_k^{+(-)}(\{n_{ij}\}_\alpha)$ is independent of the magnetic field. The magnetic field dependence of the tunneling current comes from the probability $p(\{n_{ij}\}_\alpha)$ of charge state $\{n_{ij}\}_\alpha$ which is determined by the detailed balance equation, $\mathbf{M}\mathbf{p} = 0$. Since the transition matrix \mathbf{M} contains the tunneling rates between magnetic particles, the matrix \mathbf{M} and therefore the probability $p(\{n_{ij}\}_\alpha)$ can be modified by applying the magnetic field. For the bias voltage just above the threshold voltage V_{th} , we have a few charge states contributing to the tunneling current and tunneling rates with these charge states are very different from each other due to the charging energy. Therefore, the strong modification of the probability $p(\{n_{ij}\}_\alpha)$ is made to satisfy the detailed balance equation and the TMR is strongly enhanced just above the threshold voltage V_{th} .^{3,4,12} This kind of TMR enhancement in double tunnel junctions has been studied by Barnás and Fert³ and by Majumdar and Hershfield.⁴ They also predicted the oscillating behavior of TMR against the bias voltage. We have a TMR oscillation in our system, however, the magnitude of the oscillation is small since there is no bottleneck of the tunneling in our junction array. Moreover, we have many junction arrays in the nanobridge and the randomness of junction capacitances also smears the TMR oscillation.

In order to explain the experimental results for the sample with $w = 60 \text{ nm}$, $l = 30 \text{ nm}$, and $t = 7.5 \text{ nm}$, we consider the parallel circuit of 20 triple-tunnel junctions and assume that the tunnel resistance between the electrode and a particle is expressed as $R_{ep} = (1 \pm \delta)\bar{R}_{ep}$ where δ is the deviation from the typical value \bar{R}_{ep} . Other junction parameters such as tunnel resistances between particles R_{pp} , junction capacitances C_{ep} , and C_{pp} are also assumed to be distributed around the mean values, i.e., $R_{pp} = (1 \pm \delta)\bar{R}_{pp}$, $C_{ep} = (1 \pm \delta)\bar{C}_{ep}$, and $C_{pp} = (1 \pm \delta)\bar{C}_{pp}$. The deviation δ for each junction parameter is randomly chosen within the range of $-0.1 < \delta < 0.1$. The temperature is set to be 4.2 K and the typical value of tunnel resistances for the parallel alignment of magnetizations is taken to be $\bar{R}_{pp} = \bar{R}_{ep}/2$. The

tunnel resistance between particles for the antiparallel alignment of magnetizations is larger than that for the parallel alignment and is expressed by using the spin polarization P as $\bar{R}_{pp} = (\bar{R}_{ep}/2) \cdot (1 + P^2)/(1 - P^2)$, where P is assumed to be 0.42 for Co.¹⁵ The typical values of junction capacitances are taken to be $\bar{C}_{ep} = 0.1 \text{ aF}$, and $\bar{C}_{pp} = 0.05 \text{ aF}$.¹²

The TMR obtained by the numerical calculation are shown in Fig. 3(b). One can see that the theoretical result is in good agreement with the experimental one. The TMR is enhanced just above the threshold voltage V_{th} and decreases with the bias voltage. The randomness of junction capacitances smears the oscillation of the total TMR as shown in Fig. 3(b). The difference between voltages V_{th} and V_p shown in Fig. 4 may be caused by the effects we have not considered, such as leak current through the glass substrate.

In conclusion, we have fabricated granular nanobridges consisting of electrodes separated by a nanometer-sized gap in which a thin insulating granular film is filled, and investigated the spin-dependent transport. The Coulomb blockade with a clear threshold voltage (V_{th}) is observed. TMR shows a maximum value exceeding about 30% at the V_b slightly above V_{th} . This enhancement is explained by the orthodox theory of SET in ferromagnetic multiple junctions.

The present measurements have been performed at 4.2 K , because at higher temperatures the leak current flowing through the paths besides the gap is not negligible. When the nanobridge structure is improved properly, the enhanced TMR is expected to appear even at room temperature, since the charging energy of a particle exceeds the thermal energy at room temperature. Therefore, the results obtained in this study may provide potential applications for new magnetic devices in the future.

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¹in *Single Charge Tunneling*, NATO ASI Series, Vol. 294, edited by H. Grabert and M. H. Devoret (Plenum, New York, 1992).

²S. Takahashi and S. Maekawa, Phys. Rev. Lett. **80**, 1758 (1998).

³J. Barnás and A. Fert, Phys. Rev. Lett. **80**, 1058 (1998).

⁴K. Majumdar and S. Hershfield, Phys. Rev. B **57**, 11 521 (1998).

⁵A. Brataas, Yu. V. Nazarov, J. Inoue, and G. E. Bauer, Phys. Rev. B **59**, 93 (1999).

⁶K. Ono, H. Shimada, and Y. Ootuka, J. Phys. Soc. Jpn. **66**, 1261 (1997).

⁷A. Bezryadin, C. Deller, and G. Schmid, Appl. Phys. Lett. **71**, 1273 (1997).

⁸E. M. Ford and H. Ahmed, Appl. Phys. Lett. **75**, 421 (1999).

⁹K. Nakajima, Y. Saito, S. Nakamura, and K. Inomata, IEEE Trans. Magn. (to be published).

¹⁰H. Fujimori, S. Mitani, and S. Ohnuma, Mater. Sci. Eng., B **31**, 219 (1995).

¹¹K. Yakushiji, S. Mitani, K. Takanashi, J.-G. Ha, and H. Fujimori, J. Magn. Mater. **212**, 75 (2000).

¹²H. Imamura, J. Chiba, S. Mitani, K. Takanashi, S. Takahashi, S. Maekawa, and H. Fujimori, Phys. Rev. B **61**, 46 (2000).

¹³S. Guéron, M. M. Deshmukh, E. B. Myers, and D. C. Ralph, Phys. Rev. Lett. **83**, 4148 (1999).

¹⁴S. Mitani, S. Takahashi, K. Takanashi, K. Yakushiji, S. Maekawa, and H. Fujimori, Phys. Rev. Lett. **81**, 2799 (1998).

¹⁵R. J. Soulen Jr., J. M. Byers, M. S. Osofsky, B. Nadgorny, T. Ambrose, S. F. Cheng, P. R. Broussard, C. T. Tanaka, J. Nowak, J. S. Moodera, A. Barry, and J. M. D. Coey, Science **282**, 85 (1998).